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# CRACK PROPAGATION AND DEFORMATION IN PZT TRANSDUCER CERAMICS

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This report summarizes studies completed over the period 3/1/76 to 2/29/80 representing a research program on environmental (chemical, electrical and mechanical) effects on crack propagation in the piezoelectric transducer ceramic PZT. Stress relaxation and the photomechanical effect in PZT are also discussed.

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## ABSTRACT

This report summarizes the environmental (chemical, mechanical and electrical) effects on crack propagation in the piezoelectric transducer ceramic, PZT, and also the phenomena of stress relaxation and the photomechanical effect as related to lead zirconate titanite (PZT). Slow crack growth in PZT is sensitive to both chemical and electrical testing environments. Moisture and AC or DC fields applied perpendicularly to the crack plane enhance crack propagation in unpoled PZT. In poled PZT, AC or DC fields cause the crack to deviate or turn out of its original plane.

Stress relaxation or the time-dependent deformation of PZT and the effect of light (photomechanical effect) on stress relaxation are reviewed. The stress relaxation data can be treated in terms of an analysis of thermally activated deformation.

# I. INTRODUCTION

This final report on Contract No. N00014-76-C-0625 summarizes research conducted over the past four years at Honeywell Corporate Material Sciences Center on the environmental effects on crack propagation in lead zirconate titanate, a piezoelectric transducer ceramic. An index of all Technical Reports generated during the contract and the associated publications and presentations are listed in Sections II and III. Section IV presents a brief review of our findings.

## II. INDEX OF TECHNICAL REPORTS ON CONTRACT N00014-76-C-0625

First Technical Report, "Crack Propagation in PZT," J. G. Bruce and B. G. Koepke, July 1977.

Second Technical Report, "Stress Relaxation in PZT," B. G. Koepke, K. A. Esaklul and W. W. Gerberich, October 1978.

Third Technical Report, "The Photomechanical Effect in PZT," B. G. Koepke, K. A. Esaklul and K. D. McHenry, 1980.

Fourth Technical Report, "Electrical Field Effects on Crack Propagation in PZT," K. D. McHenry and B. G. Koepke, May 1980.

Fifth Technical Report, "Identation Fracture of PZT," K. D. McHenry and B. G. Koepke, June 1980.

## III. PUBLICATIONS AND PRESENTATION ON CONTRACT N00014-76-C-0625

PUBLICATIONS

1. J. G. Bruce and B. G. Koepke, "Evaluation of $K_{IC}$ by the Double-Torsion Technique," J. Amer. Ceram. Soc., 60, 284 (1977).

2. J. G. Bruce, W. W. Gerberich and B. G. Koepke, "Subcritical Crack Growth in PZT," Proceedings of the International Symposium on Fracture Mechanics of Ceramics, Vol. 4, p. 687, Plenum Press (1978).

3. B. J. Pletka, E. R. Fuller, Jr. and B. G. Koepke, "An Evaluation of Double Torsion Testing - Experimental," Proceedings of the ASTM Symposium on Fracture Mechanics Tests for Brittle Materials, ASTM STP 678, p. 19 (1979).

4. K. A. Esaklul, W. W. Gerberich and B. G. Koepke, "Stress Relaxation in PZT," J. Amer. Ceram. Soc., 63, 25 (1980).

5. B. G. Koepke and K. D. McHenry, "Fracture and Deformation of PZT," Ferro-electrics, 28, 343 (1980).

6. K. A. Esaklul, B. G. Koepke and K. D. McHenry, "The Photomechanical Effect in PZT," to be submitted to J. Amer. Ceram. Soc.

7. K. D. McHenry and B. G. Koepke, "Electric Field Effects on Crack Propagation in PZT," to be submitted to J. Amer. Ceram. Soc.

8. K. D. McHenry and B. G. Koepke, "Indentation Fracture of PZT and $BaTiO_3$," to be submitted to J. Amer. Ceram. Soc.

PRESENTATIONS

1. B. G. Koepke and R. G. Johnson, "Crack Propagation in PZT," Paper presented at American Ceramic Society Annual Meeting, Cincinnati, May 1976.

2. B. G. Koepke and R. G. Johnson, "The Effects of Electrical and Chemical Environment on Fracture of PZT High Drive Sonar Ceramics," Paper presented at Australian Workshop on Sonar Materials by Dr. A. Diness of the Office of Naval Research, July 1976.

3. R. G. Johnson, S. J. Tibbetts, J. G. Bruce and B. G. Koepke, "Effects of AC and DC Electric Fields on Crack Propagation in PZT," Paper presented at American Ceramic Society Annual Meeting, Chicago, April 1977.

4. J. G. Bruce and B. F. Koepke, "En ironmental Effects and the Mechanism of Fracture During Slow Crack Growth in PZT," Paper presented at American Ceramic Society Annual Meeting, Chicago, April 1977.

5. J. G. Bruce, W. W. Gerberich and B. G. Koepke, "Subcritical Crack Growth in PZT," Paper presented at International Symposium on Fracture Mechanics of Ceramics, Pennsylvania State University, July 1977.

6. B. G. Koepke, "Experimental Aspects of the Double Torsion Test," Paper presented at ASTM Subcommittee E24.07 (Fracture Testing of Brittle Non-Metallic Materials) meeting, NBS, October 1977.

7. B. G. Koepke, "Environmental Effects on Fracture of PZT," Paper presented at Joint Materials Science Physics Colloquium, Univ. of Kansas, November 1977.

8. K. A. Esaklul and B. G. Koepke, "Photomechanical Effects in PZT," Paper presented at American Ceramic Society Annual Meeting, Detroit, May 1978.

9. B. J. Pletka, E. R. Fuller, Jr. and B. G. Koepke, "An Evaluation of Double Torsion Testing-Experimental," Paper presented at ASTM Symposium on Fracture Mechanics Tests for Brittle Materials, Virginia Polytechnic Institute, June 1978.

10. B. G. Koepke, K. A. Esaklul and W. W. Gerberich, "Stress Relaxation in PZT," Paper presented at the Fall Meeting of the American Ceramic Society Basic Science Division, NBS, November 1978.

11. J. M. Smeby, K. D. McHenry and B. G. Koepke, "Indentation Fracture of PZT and $BaTiO_3$," Paper presented at American Ceramic Society Annual Meeting, Cincinnati, April 1979.

12. R. A. Munson, K. D. McHenry and B. G. Koepke, "Electric Field Effects on Crack Propagation in PZT," Paper presented at American Ceramic Society Annual Meeting, Cincinnati, April 1979.

13. B. G. Koepke and K. D. McHenry, "Fracture and Deformation of PZT," Paper presented at IEEE Symposium on Applications of Ferroelectrics, Minneapolis, June 1979.

14. K. D. McHenry, T. E. Nohava and B. G. Koepke, "Electric Field Effects on Crack Propagation in Poled PZT Transducer Ceramics," Paper presented at American Ceramic Society Annual Meeting, Chicago, April 1980.

# IV. CRACK PROPAGATION AND DEFORMATION IN PZT TRANSDUCER CERAMICS

The propagation of surface and subsurface flaws under subcritical loads in ceramic high-drive sonar transducers can be detrimental to both the mechanical strength and the electrical characteristics of a device. The phenomenon of subcritical crack growth has been studied in many ceramic materials[1] but has only recently been investigated in transducer ceramics. Most of the work has concentrated on the effects of chemical environments on subcritical crack growth in transducer ceramics. Little or no work has been conducted to investigate the effects of applied electric fields on subcritical crack growth in these materials. In this report, we summarize the results of our four-year investigation into the chemical and electrical effects on subcritical crack growth in a lead zirconate titanate (PZT). The PZT used throughout this investigation was manufactured by the Honeywell Ceramics Center and has a nominal composition of $Pb_{0.94}Sr_{0.06}Ti_{0.47}Zr_{0.53}O_3$ plus proprietary additions. This composition lies just to the right of the morphotropic phase boundary in the tetragonal phase field.

We also summarize the results of our findings on the time-dependent deformation and the photomechanical effect in PZT. When piezoelectric ceramics are subjected to an applied stress, a large part of the resulting strain is time-dependent and reversible. This anelastic component of the deformation is enhanced stress relaxation under the influence of illumination. Studies of the relation of domain processes to deformation in piezoelectric ceramics have practical significance due to the relaxation between deformation, domain processes and aging; i.e., the degradation of the properties of a poled specimen with time.

## A. CHEMICAL AND TEMPERATURE EFFECTS ON SUBCRITICAL CRACK GROWTH IN PZT

All subcritical crack growth results in this investigation were taken using the double torsion testing technique popularized by Evans et. al.[2,3]. In the portion of the investigation described here, the double torsion specimen was tested in the fixed grip mode utilizing the load relaxation technique.

Subcritical crack growth in PZT was found to be sensitive to moisture. Specimens were tested in mineral oil, freon, toluene and distilled water. Mineral oil and freon are essentially inert environments with respect to moisture while toluene contains several ppm $H_2O$. Slow crack growth was evident in all environments but crack growth was enhanced with moisture content. In mineral oil and freon, the slope of the ($V$-$K_I$) crack velocity-stress intensity factor data was $\approx$ 130, indicating little or no susceptibility to slow crack growth in these environments. However, the slope of the ($V$-$K_I$) data in water was <50 indicating a very marked increase in the material's susceptibility to slow crack growth in this environment. In addition, the ($V$-$K_I$) data in water was shifted to lower $K_I$ values as compared to mineral oil or freon environments. Thus, the overall effect is that for a given applied stress, i.e., stress intensity, cracks will propagate at much higher velocities in moisture-containing environments than in inert environments.

To examine the effects of temperature on crack propagation in PZT, ($V$-$K_I$) curves were determined on unpoled samples tested in distilled water at $0^\circ$, $25^\circ$, $50^\circ$ and $75^\circ$C and in mineral oil at $25^\circ$, $75^\circ$, $100^\circ$ and $125^\circ$C. The overall effect of increasing temperature is to increase crack propagation in PZT. The data from tests

performed in water were analyzed in terms of a thermally activated process under the assumption that a single process is rate controlling. The resultant analysis yielded a stress-free activation energy of 100kcal/mole.

## B. AC AND DC FIELD EFFECTS ON SLOW CRACK GROWTH IN UNPOLED PZT

In order to facilitate the fracture mechanics investigation of piezoelectric materials subjected to electric fields, a dead-weight loading system was developed. With this technique, a fixed load is applied to the specimen and the crack velocity is determined by monitoring the deflection rate of the specimen with an LVDT. This system was necessitated by the fact that the large piezoelectric effect in PZT, i.e., the large piezoelectric shape deformations, eliminated the possibility of using the fixed grip or *constant displacement* testing technique.

Electric fields were applied both parallel and perpendicular to the crack plane. Fields applied parallel to the crack plane had no noticeable effect on crack growth under any testing condition. The effect of AC or DC fields applied perpendicular to the crack plane was to accelerate crack growth with increasing field strengths (1-4 KV/cm). AC fields were applied at a constant frequency of 60 Hz. While both AC and DC fields served to enhance slow crack growth in PZT, the dependence of the slow crack growth data on a 1 KV/cm DC field versus 1 KV/cm 60 Hz AC field, for example, was not exactly equivalent. The difference in the functional dependence of the crack growth data for equal AC and DC field strengths must then be a result of the time-dependent nature of the applied AC field.

The frequency of an applied AC field may also have a deleterious effect in increasing the crack growth in PZT, providing the frequency of the applied field matches a mechanical resonance or harmonic of the test specimen. However, it should be noted that this frequency is continually changing with crack growth. In general, as the crack becomes longer, the mechanical resonance frequency of the specimen decreases. The frequency of the applied AC field must then be continually adjusted in order to match the resonant frequency.

When high DC fields (∿8KV/cm) are applied perpendicularly to the crack plane in PZT, the crack turns out of its original plane and propagates toward the side of the double torsion specimen. In addition, the cracks always turn in a direction against the applied mode fracture via the introduction of shear components to the mechanical stress field at the crack tip. Fractography reveals a distortion of the original crack tip profile in these specimens and supports a mixed mode fracture hypothesis.

## C. AC AND DC FIELD EFFECTS ON SLOW CRACK GROWTH IN POLED PZT

Composite double torsion specimens were developed to study the effects of electric fields on subcritical crack growth in poled PZT. The development of these specimens aided in reducing the poling fields necessary for the polarization of the material. The central sections of the double torsion specimens were cut out, and only these sections were poled. These sections were ≈0.5cm in width. These pieces were subsequently poled at 150°C in peanut oil with an applied field of 20 KV/cm. The poled sections were then rejoined with the original double torsion

pieces by silver conducting epoxy. The poling direction was perpendicular to the crack plane.

The effect of AC or DC fields applied perpendicularly to the crack plane in the poled specimens was to turn the crack out of its original plane toward the side of the specimen. In the case of the poled material, DC fields applied parallel or antiparallel to the poling direction always turn the crack in a direction against the original poling direction.

These results are consistent with those obtained for unpoled material in terms of which way the crack tends to turn. In the case of poled material, the crack always turns against the polarization direction. In unpoled PZT, the crack turns against the applied field direction. However in unpoled PZT, the applied field direction also is the polarization direction. The overall effect then is for cracks to turn against the intended or induced polarization direction. However, an important point should be noted. In order for the crack to turn, some complex interaction between the applied field and the polarization is required. The previous statement on the interaction of the field and polarization is based on two observations:

- In unpoled PZT, small applied fields (<4 KV/cm) accelerate crack growth but do not turn the crack.
- In poled PZT, when a field is not applied, the cracks do not turn, but rather run down the middle of the specimen.

The origin of shear components causing the crack to turn is a result of the interaction of the piezoelectric $d_{15}$ coefficient

from tetragonal 4mm symmetry and off-axis components of the applied electric field. Off-axis components of the electric field occur because of the field lines curving around the crack tip. The directionality of the crack deviation from its original plane is dictated by the polarization direction. This phenomenon is not clearly understood, but the directionality may be influenced by the depolarization field within the material.

## D. FRACTURE TOUGHNESS OF PZT

The fracture toughness, $K_{IC}$, or resistance to rapid crack propagation was measured using three different techniques. These were the double torsion, the controlled surface flaw, and the indentation fracture techniques. The fracture toughness of PZT as de-determined by the double torsion technique was 0.75 $MNm^{-3/2}$, regardless of testing environment (chemical or electrical).

With the controlled surface flaw technique, indentation loads of >20 lbs, utilizing the Knoop diamond indent, were required to yield a satisfactory semi-elliptical flaw. The fracture toughness was calculated from the fracture stress and the measured flaw size. In all cases, the fracture toughness as measured by the controlled surface flaw technique was equivalent to that measured by the double torsion technique. For many ceramics, it is necessary to remove several microns of material following indentation in order to eliminate residual surface stresses. This was not the case for PZT, due either to densification or stress relaxation as a result of domain boundary migration. Either of these processes would hinder the development of a residual surface stress.

The indentation fracture technique was found not to be suitable for determining the true value of $K_{IC}$ in PZT. With this technique, a Vicker's indentation is made on the surface of the material and $K_{IC}$ calculated from the ratio of the radial crack lengths to the indent radius. For PZT, this technique yielded apparent $K_{IC}$ values two to three times larger than either of the other two techniques.

The lengths of the radial cracks that formed from the corners of the indent impression were much shorter than predicted lengths, assuming a $K_{IC} = 0.75 \text{ MNm}^{-3/2}$. This result can be explained by noting that in PZT several energy dissipative mechanisms are available other than crack formation. One is densification, and the other is domain boundary migration. Thus, the fracture toughness of the material measured with this technique is artificially high, since some of the energy has been dissipated by mechanisms other than crack formation and extension.

## E. STRESS RELAXATION IN PZT

Stress relaxation in PZT has been found to occur at relatively low values of stress and to depend systematically on both stress and temperature. The stress and temperature dependence of the relaxation behavior suggest that the deformation mechanism is thermally activated. Logarithmic stress relaxation is observed only over a fraction of the time intervals used in the tests, and the data cannot be analyzed in terms of the unusual Arrhenius equation. An analysis based on the fundamental expression in reaction rate theory, which allows for forward and reverse activation of the deformation mechanism, was successfully applied

to the results; an experimental activation energy and activation volume of 25 kcal/mole and 3,000 to 7,000Å respectively were measured.

The accepted deformation mechanism for piezoelectric ceramics is non-180-degree domain boundary migration. A dislocation model for non-180-degree domain boundary (twin boundary) migration has been developed that is consistent with the experimental activation parameters. The data strongly suggest that domain boundary migration in PZT occurs by the homogeneous nucleation of twinning dislocation loops on lattice planes immediately adjacent to the boundary.

## F. PHOTOMECHANICAL EFFECTS IN PZT

During the course of this investigation, the photomechanical effect in PZT was discovered in our laboratory. The overall effect of illumination is to enhance stress relaxation in PZT. In addition, the stress relaxation is a function of the intensity of the illumination and temperature as discussed previously. This phenomenon cannot be modeled simply in terms of thermally activated deformation as was done for stress relaxation. The activation analysis indicates that illumination is also part of the activation event and must be accounted for. Clearly, more work is required for a total understanding of the photomechanical effect in PZT.

## REFERENCES


1. A. G. Evans and T. G. Langdon, Prog. Mat. Sci., 21, 171 (1976).

2. A. G. Evans, J. Mater, Sci., 7, 1137 (1972).

3. D. P. Williams and A. G. Evans, J. Testing and Eval., 1, 264 (1973).